

# Thermal Conductivity of the Quasi One-Dimensional Spin System $\text{Sr}_2\text{V}_3\text{O}_9$

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We have measured the thermal conductivity along the  $[101]$  direction,  $\kappa_{[101]}$ , along the  $[10\bar{1}]$  direction,  $\kappa_{[10\bar{1}]}$ , and along the  $b$ -axis,  $\kappa_b$ , of the quasi one-dimensional  $S=1/2$  spin system  $\text{Sr}_2\text{V}_3\text{O}_9$  in magnetic fields up to 14 T, in order to find the thermal conductivity due to spinons and to clarify whether the spin-chains run along the  $[101]$  or  $[10\bar{1}]$  direction. It has been found that both  $\kappa_{[101]}$ ,  $\kappa_{[10\bar{1}]}$  and  $\kappa_b$  show one peak around 10 K in zero field and that the magnitude of  $\kappa_{[10\bar{1}]}$  is larger than those of  $\kappa_{[101]}$  and  $\kappa_b$ . By the application of magnetic field along the heat current, the peak of  $\kappa_{[10\bar{1}]}$  is markedly suppressed, while the peaks of  $\kappa_{[101]}$  and  $\kappa_b$  little change. These results indicate that there is a large contribution of spinons to  $\kappa_{[10\bar{1}]}$  and suggest that the spin-chains run along the  $[10\bar{1}]$  direction.

## I. INTRODUCTION

Recently, the thermal conductivity in low-dimensional quantum spin systems has attracted interest, because a large contribution of magnetic excitations to the thermal conductivity has been observed in some compounds regarded as low-dimensional quantum spin systems. In one-dimensional (1D) antiferromagnetic (AF) Heisenberg spin systems with the spin quantum number  $S = 1/2$ , especially, it has theoretically been proposed that the thermal conduction due to magnetic excitations, namely, spinons is ballistic [1–3]. In fact, a large contribution of spinons to the thermal conductivity has been observed in the  $S=1/2$  1D AF Heisenberg spin system  $\text{Sr}_2\text{CuO}_3$  [4]. Moreover, the ballistic nature of the thermal conduction due to spinons has experimentally been confirmed in  $\text{Sr}_2\text{CuO}_3$  [5].

The compound  $\text{Sr}_2\text{V}_3\text{O}_9$  contains three kinds of vanadium ions in the unit cell. Two of them are nonmagnetic  $\text{V}^{5+}$  ions located in  $\text{VO}_4$  tetrahedra, and the rest is  $\text{V}^{4+}$  ions with  $S = 1/2$  located in  $\text{VO}_6$  octahedra. The  $\text{VO}_6$  octahedra are connected with each other by sharing an oxygen at the corner along the  $[101]$  direction, as shown Fig. 1. The  $ac$ -plane with the magnetic network is weakly stacked along the  $b$ -axis. The magnetic properties are understood in terms of the  $S=1/2$  1D AF chain model with the exchange interaction between the nearest spins,  $J = 82$  K, estimated from the suscepti-

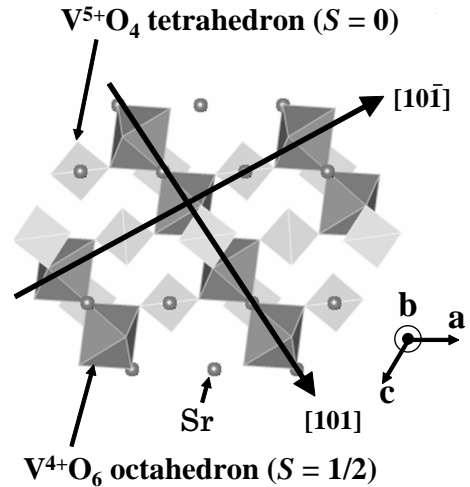


FIG. 1: Crystal structure of  $\text{Sr}_2\text{V}_3\text{O}_9$ .  $\text{V}^{5+}$  ions and  $\text{V}^{4+}$  ions are located in  $\text{VO}_4$  tetrahedra and  $\text{VO}_6$  octahedra, respectively.  $\text{VO}_6$  octahedra are connected with each other along the  $[101]$  direction by sharing an oxygen at the corner and are also connected via a  $\text{VO}_4$  tetrahedron along the  $[10\bar{1}]$  direction.

bility measurements [6]. However, it has been suggested from the ESR measurements [7] and a theory by Koo and Whangbo [8] that the spin-chain direction is not the  $[101]$  direction but the  $[10\bar{1}]$  direction, along which  $\text{VO}_6$  octahedra are connected via a  $\text{VO}_4$  tetrahedron as shown Fig. 1.

Therefore, in order to find the contribution of spinons

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to the thermal conductivity and also to clarify whether the spin-chain direction is the  $[101]$  or  $[10\bar{1}]$  direction, we have measured the thermal conductivity of  $\text{Sr}_2\text{V}_3\text{O}_9$  along the  $[101]$ ,  $[10\bar{1}]$  directions and the  $b$ -axis. The precise measurement of thermal conductivity needs a large-size single crystal. Therefore, we have attempted to grow large-size single crystals of  $\text{Sr}_2\text{V}_3\text{O}_9$  by the floating-zone (FZ) method.

## II. EXPERIMENTAL

First, polycrystalline powder of  $\text{Sr}_2\text{V}_2\text{O}_7$  was prepared by the solid-state reaction method, in order to prepare a feed rod for the FZ growth. The prescribed amount of  $\text{SrCO}_3$  and  $\text{V}_2\text{O}_5$  powder was mixed in the molar ratio of  $\text{SrCO}_3 : \text{V}_2\text{O}_5 = 2 : 1$  and pre-fired in air at  $700^\circ\text{C}$  for 72 h. After pulverization, the pre-fired powder of  $\text{Sr}_2\text{V}_2\text{O}_7$  was mixed with  $\text{VO}_2$  powder in the molar ratio of  $\text{Sr}_2\text{V}_2\text{O}_7 : \text{VO}_2 = 1 : 1$  and isostatically cold-pressed at 600 bar into a rod of 7 mm in diameter and  $\sim 100$  mm in length. Then, the rod was sintered at  $540^\circ\text{C}$  in Ar for 24 h. As a result, a sintered feed rod was prepared. The FZ growth was carried out by the twice-scanning technique in an infrared heating furnace equipped with a double ellipsoidal mirror (NEC Machinery Corp, Model SC-K15HD-H). A high-density premelted feed rod was prepared through the first scan using the sintered feed rod. In the first scan, the molten zone was scanned at a speed of  $\sim 20$  mm/h under flowing Ar of 1.5 atm. Next, the second scan, namely, a usual growing procedure was carried out using the premelted feed rod at the growth rate of 1.0 mm/h in the same atmosphere as in the first scan. Thermal-conductivity measurements were carried out by the conventional steady-state method.

## III. RESULTS AND DISCUSSION

We have succeeded in growing a single-crystal rod, owing to the stable upkeep of the molten zone during the FZ growth. Figure 2(a) shows an as-grown single-crystal rod with  $\sim 6$  mm in diameter and  $\sim 100$  mm in length. The grown crystals were characterized by the x-ray back-Laue photography, as shown in Fig. 2(b). Although the grown crystals were composed of several domains, the diffraction spots were very sharp. The dimensions of the single-domain region were typically  $\sim 6$  mm in diameter and  $\sim 25$  mm in length. The single crystals were also confirmed by the powder x-ray diffraction to be of the single phase without any impurity phases. Accordingly, it is concluded that we have succeeded in the growth of high-quality single-crystals. The high quality was supported by the magnetic-susceptibility result that no Curie term due to impurities and/or lattice defects was observed at very low temperatures.

Figure 3 shows the temperature dependence of the thermal conductivity along the  $[101]$  direction,  $\kappa_{[101]}$ ,

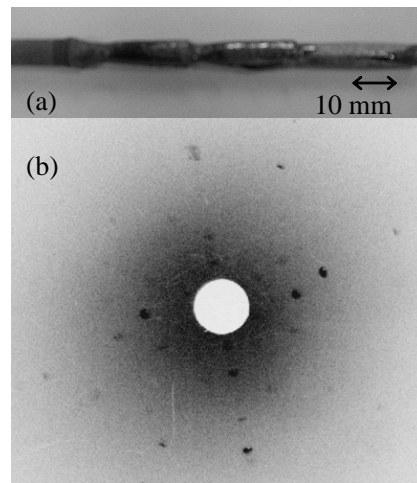


FIG. 2: (a) Picture of an as-grown single-crystal rod of  $\text{Sr}_2\text{V}_3\text{O}_9$ . (b) X-ray back-Laue photograph of an as-grown single-crystal in the x-ray parallel to the  $b$ -axis.

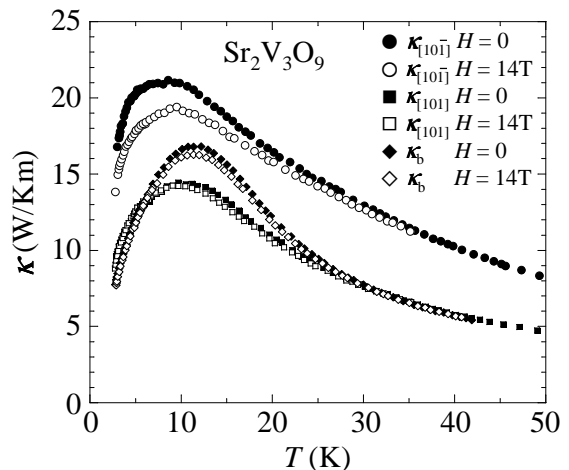


FIG. 3: Temperature dependence of the thermal conductivity of  $\text{Sr}_2\text{V}_3\text{O}_9$  along the  $[101]$  direction,  $\kappa_{[101]}$ , along the  $[10\bar{1}]$  direction,  $\kappa_{[10\bar{1}]}$ , and along the  $b$ -axis,  $\kappa_b$ , in zero field and a magnetic field of 14 T parallel to the respective heat current.

along the  $[10\bar{1}]$  direction,  $\kappa_{[10\bar{1}]}$ , and along the  $b$ -axis,  $\kappa_b$ , in zero field and a magnetic field of 14 T parallel to the respective heat current. In zero field, both  $\kappa_{[101]}$ ,  $\kappa_{[10\bar{1}]}$  and  $\kappa_b$  show a peak around 10 K. The magnitude of  $\kappa_{[10\bar{1}]}$  at the peak is larger than those of  $\kappa_{[101]}$  and  $\kappa_b$ . Since  $\text{Sr}_2\text{V}_3\text{O}_9$  is an insulator, the thermal conductivity is described as the sum of the thermal conductivity due to phonons,  $\kappa_{\text{phonon}}$ , and due to spinons,  $\kappa_{\text{spinon}}$ . It is known that the anisotropy of  $\kappa_{\text{phonon}}$  is usually not so large and that the contribution of  $\kappa_{\text{spinon}}$  markedly appears along the direction where the magnetic interaction is strong. Therefore, the large anisotropy of the thermal conductivity is guessed to be due to a large contribution

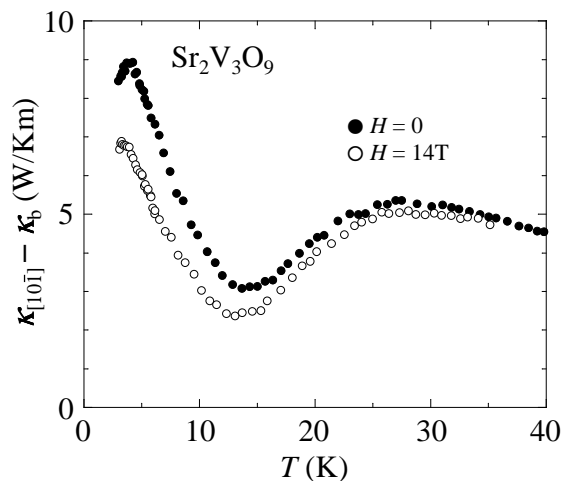


FIG. 4: Temperature dependence of the difference between  $\kappa_{[101]}$  and  $\kappa_b$  in zero field and 14 T.

of  $\kappa_{\text{spinon}}$  to  $\kappa_{[101]}$ . By the application of magnetic field parallel to the heat current, the peak of  $\kappa_{[101]}$  around 10 K is suppressed with increasing field, while there is little change in  $\kappa_{[101]}$  and  $\kappa_b$ , as shown in Fig. 3. This result also supports the guess that there is a large contribution of  $\kappa_{\text{spinon}}$  to  $\kappa_{[101]}$ , because  $\kappa_{\text{spinon}}$  is expected to be affected by the application of a magnetic field comparable with  $J/(g\mu_B)$  ( $g$ : the  $g$ -factor,  $\mu_B$ : the Bohr magneton). Furthermore, the little change in  $\kappa_{[101]}$  and  $\kappa_b$  by the application of magnetic field indicates that the contribution of  $\kappa_{\text{spinon}}$  is very small along the  $[101]$  direction and  $b$ -axis. Accordingly, it is concluded that the spin-chain direction is the  $[10\bar{1}]$  direction, as suggested from the ESR measurements [7] and the theory by Koo and Whangbo [8].

Here, we estimate  $\kappa_{\text{spinon}}$  in  $\kappa_{[10\bar{1}]}$ , where both  $\kappa_{\text{spinon}}$  and  $\kappa_{\text{phonon}}$  are included. In the temperature dependence of  $\kappa_{[10\bar{1}]}$ , only one peak is observed around 10 K, indicating that both peaks of  $\kappa_{\text{spinon}}$  and  $\kappa_{\text{phonon}}$  are overlapped. Therefore, it is very hard to estimate  $\kappa_{\text{spinon}}$  and  $\kappa_{\text{phonon}}$  separately. As for  $\kappa_{[101]}$ , a small contribution of  $\kappa_{\text{spinon}}$  to  $\kappa_{[101]}$  is guessed to exist, because the  $[101]$  direction is not exactly perpendicular to the  $[10\bar{1}]$  direction but

$84.48^\circ$  tilted from the  $[10\bar{1}]$  direction. As for  $\kappa_b$ , it is due to only  $\kappa_{\text{phonon}}$ . Therefore, neglecting the anisotropy of  $\kappa_{\text{phonon}}$ ,  $\kappa_{\text{spinon}}$  along the  $[10\bar{1}]$  direction is very roughly estimated to be  $\kappa_{[10\bar{1}]} - \kappa_b$ , as shown in Fig. 4. However, this is not simply accepted as the temperature dependence of  $\kappa_{\text{spinon}}$  along the  $[10\bar{1}]$  direction, because unusual two peaks appear around 4 K and 28 K. What is remarkable at least is that the peak around 4 K is strongly suppressed by the application of magnetic field while the other peak around 28 K little changes. Therefore, it is expected that the peak around 4 K is attributed to the contribution of  $\kappa_{\text{spinon}}$ . On the other hand, it is likely that the peak around 28 K appears because of the difference of  $\kappa_{\text{phonon}}$  between the  $[10\bar{1}]$  direction and the  $b$ -axis. Accordingly, at least these results indicate that the temperature dependence of  $\kappa_{\text{spinon}}$  exhibits a peak around 4 K in  $\kappa_{[10\bar{1}]}$ . In order to estimate the value of  $\kappa_{\text{spinon}}$  in  $\text{Sr}_2\text{V}_3\text{O}_9$  exactly, the estimate of the anisotropy of  $\kappa_{\text{phonon}}$  between the  $[10\bar{1}]$  direction and  $b$ -axis is necessary.

#### IV. SUMMARY

Large-size single-crystals of  $\text{Sr}_2\text{V}_3\text{O}_9$  have successfully been grown by the FZ method and the thermal conductivity have been measured in magnetic fields up to 14 T. The magnitude of  $\kappa_{[10\bar{1}]}$  in zero field is larger than those of  $\kappa_{[101]}$  and  $\kappa_b$ . By the application of magnetic field, only  $\kappa_{[10\bar{1}]}$  is suppressed. These anisotropic behaviors suggest that the spin-chains run along the  $[10\bar{1}]$  direction. Moreover, it is concluded from the field effect of  $\kappa_{[10\bar{1}]} - \kappa_b$  related to the behavior of  $\kappa_{\text{spinon}}$  that the temperature dependence of  $\kappa_{\text{spinon}}$  exhibits a peak around 4 K.

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